10/530002
JC17 Rec'd PCT/PTO 31 MAR 2005

Amendments to the Claims

Please cancel Claims 1-28. Please add new Claims 29-58. The Claim Listing below will replace all prior versions of the claims in the application:

Claim Listing

1-28 (Cancelled)

29. (New) A proton-conducting polymer membrane which comprises polyazoles containing phosphonic acid groups and is obtainable by a process comprising the steps

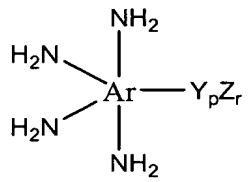
A) mixing one or more aromatic or heteroaromatic tetraamino compounds with one or more aromatic or heteroaromatic carboxylic acids or derivatives thereof which contain at least two acid groups per carboxylic acid monomer, with at least part of the tetraamino compounds or the carboxylic acids comprising at least one phosphonic acid group, or mixing of one or more aromatic or heteroaromatic diaminocarboxylic acids, of which at least part comprises phosphonic acid groups, in polyphosphoric acid to form a solution or dispersion,

B) heating the solution or dispersion obtained according to step A) under inert gas to temperatures of up to 350°C to form polyazole polymers,

C) applying a layer using the mixture from step A) or B) to a support, and

D) treating the membrane from step C) until it is self-supporting.

30. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic tetraamino compounds of the formula (A)



(A)

where

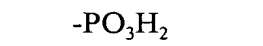
Ar is an aromatic or heteroaromatic group,

Y is a bond or a group having from 1 to 20 carbon atoms,

p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

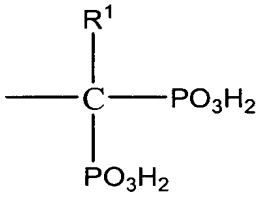
r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z is a group of the general formula (1)



(1)

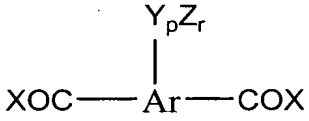
or the general formula (2)



(2)

where R^1 is a hydrogen atom or a group having from 1 to 20 carbon atoms.

31. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic carboxylic acids of the formula (B)



(B)

where

Ar is an aromatic or heteroaromatic group,

X is a halogen atom or a group of the formula OR^2, where R^2 is a hydrogen atom or a group having from 1 to 20 carbon atoms,

Y is a bond or a group having from 1 to 20 carbon atoms,

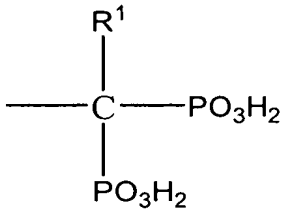
p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z is a group of the general formula (1)

$$-PO_3H_2 \qquad (1)$$

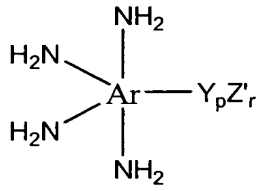
or the general formula (2)



(2)

where R^1 is a hydrogen atom or a group having from 1 to 20 carbon atoms.

32. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic tetraamino compounds of the formula (C)



(C)

where

Ar is an aromatic or heteroaromatic group,

Y is a bond or a group having from 1 to 20 carbon atoms,

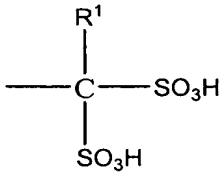
p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z' is a group of the general formula (3)

$$-SO_3H \qquad (3)$$

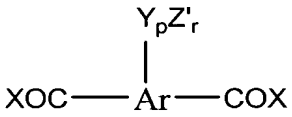
or the general formula (4)



(4)

where R^1 is a hydrogen atom or a group having from 1 to 20 carbon atoms.

33. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic carboxylic acids of the formula (D)



(D)

where

Ar is an aromatic or heteroaromatic group which optionally bears further substituents,

X is a halogen atom or a group of the formula OR^2, where R^2 is a hydrogen atom or a group having from 1 to 20 carbon atoms,

Y is a bond or a group having from 1 to 20 carbon atoms,

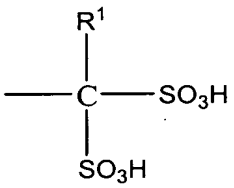
p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z' is a group of the general formula (3)

$$-SO_3H \qquad (3)$$

or the general formula (4)



(4)

where R^1 is a hydrogen atom or a group having from 1 to 20 carbon atoms.

34. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic tetraamino compounds which contain no phosphonic acid groups and aromatic or heteroaromatic carboxylic acids which contain at least one phosphonic acid group.

35. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises 3,3′,4,4′-tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine, or 1,2,4,5-tetraaminobenzene.

36. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises isophthalic acid, terephthalic acid, phthalic acid, 5-hydroxyisophthalic acid, 4-hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,N-dimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid, 2,5-dihydroxyisophthalic aid, 2,3-dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5-fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic acid, tetrafluoroterephthalic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, bis(4-carboxyphenyl) ether, benzophenone-4,4′-dicarboxylic acid, bis(4-dicarboxyphenyl) sulfone, biphenyl-4,4′-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis(4-carboxyphenyl)hexafluoropropane, 4,4′-stilbenedicarboxylic acid, 4-carboxycinnamic acid, or their C1-C20-alkyl esters or C5-C12-aryl esters, or their acid anhydrides or acid chlorides.

37. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises 2,3-diamino-5-carboxyphenylphosphonic acid, 2,3-diamino-6-carboxyphenylphosphonic acid, and 3,4-diamino-6-carboxyphenylphosphonic acid.

38. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises 2,3-diamino-5-carboxyphenylsulfonic acid, 2,3-diamino-6-carboxyphenylsulfonic acid, and 3,4-diamino-6-carboxyphenylsulfonic acid.

39. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic tricarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides or tetracarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides.

40. (New) The membrane of Claim 39, characterized in that the mixture prepared in step A) comprises 1,3,5-benzenetricarboxylic acid (trimesic acid); 2,4,5-benzenetricarboxylic acid (trimellitic acid); (2-carboxyphenyl)iminodiacetic acid, 3,5,3′-biphehyltricarboxylic acid; 3,5,4′-biphenyltricarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzene-1,2,4,5-tetracarboxylic acid; naphthalene-1,4,5,8-tetracarboxylic acid, 3,5,3′,5′-biphenyltetracarboxylic acid, benzophenonetetracarboxylic acid, 3,3′,4,4′-biphenyltetracarboxylic acid, 2,2′,3,3′-biphenyltetracarboxylic acid, 1,2,5,6-naphthalenetetracarboxyllic acid or 1,4,5,8-naphthalenetetracarboxylic acid.

41. (New) The membrane of Claim 39, characterized in that the content of tricarboxylic acid or tetracarboxylic acids is in the range of from 0 to 30 mol% based on dicarboxylic acid used.

42. (New) The membrane of Claim 41, characterized in that the content of tricarboxylic acid or tetracarboxylic acids is in the range of from 0.1 to 20 mol% based on dicarboxylic acid used.

43. (New) The membrane of Claim 42, characterized in that the content of tricarboxylic acid or tetracarboxylic acids is in the range of from 0.5 to 10 mol% based on dicarboxylic acid used.

44. (New) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises heteroaromatic dicarboxylic acids, tricarboxylic acids or tetracarboxylic

acids which contain at least one nitrogen, oxygen, sulfur, or phosphorus atom in the aromatics.

45. (New) The membrane of Claim 44, characterized in that pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid, 3,5-pyrazoledicarboxylic acid, 2,6-pyrimidinedicarboxylic acid, 2,5-pyrazinedicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid or their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid chlorides are used.

46. (New) The membrane as claimed in claim 29, characterized in that the mixture prepared in step A) comprises diaminobenzoic acid or its monohydrochloride and dihydrochloride derivatives.

47. (New) The membrane of Claim 29, characterized in that the heating according to step B) is carried our after the formation of a sheet-like structure according to step C).

48. (New) The membrane as claimed in claim 29, characterized in that the solution produced in step A) or step B) further comprises dispersed or suspended polymer.

49. (New) The membrane of Claim 29, characterized in that the treatment according to step D) is carried out at temperatures in the range of from 0°C to 150°C in the presence of moisture.

50. (New) The membrane of Claim 29, characterized in that the treatment of the membrane in step D) is carried out from 10 seconds to 300 hours.

51. (New) The membrane of Claim 29, characterized in that the membrane formed after step D) is crosslinked by action of oxygen.

52. (New) The membrane of Claim 29, characterized in that a layer having a thickness of from 20 to 4000 μm is produced in step C).

53. (New) The membrane of Claim 29, characterized in that the membrane formed after step D) has a thickness of from 15 to 3000 μm.

54. (New) An electrode having a proton-conducting polymer coating which is based on polyazoles and is obtainable by a process comprising the steps

A) mixing one or more aromatic or heteroaromatic tetraamino compounds with one or more aromatic or heteroaromatic carboxylic acids or derivatives thereof which contain at least two acid groups per carboxylic acid monomer, with at least part of the tetraamino compounds or the carboxylic acids comprising at least one phosphonic acid group, or mixing of one or more aromatic or heteroaromatic diaminocarboxylic acids, of which at least part comprises phosphonic acid groups, in polyphosphoric acid to form a solution or dispersion,

B) heating the solution or dispersion obtained according to step A) under inert gas to temperatures of up to 350°C to form the polyazole polymer,

C) applying a layer using the mixture from step A) or B) to an electrode,

D) treating the membrane formed in step C) until it has a surface hardness.

55. (New) The electrode of Claim 54, wherein the coating has a thickness of from 2 to 3000 μm.

56. (New) A membrane-electrode unit comprising at least one electrode and at least one membrane as claimed in Claim 29.

57. (New) A membrane-electrode unit comprising

at least one electrode having a proton-conducting polymer coating which is based on polyazoles and is obtainable by a process that includes the steps

A) mixing one or more aromatic or heteroaromatic tetraamino compounds with one or more aromatic or heteroaromatic carboxylic acids or derivatives thereof which contain at least two acid groups per carboxylic acid monomer, with at least part of the tetraamino compounds or the carboxylic acids comprising at least one phosphonic acid group, or mixing of one or more aromatic or heteroaromatic

diaminocarboxylic acids, of which at least part comprises phosphonic acid groups, in polyphosphoric acid to form a solution or dispersion,

B) heating the solution or dispersion obtained according to step A) under inert gas to temperatures of up to 350°C to form the polyazole polymer,

C) applying a layer using the mixture from step A) or B) to an electrode,

D) treating the membrane formed in step C) until it has a surface hardness., and

at least one membrane as claimed in Claim 29.

58. (New) A fuel cell comprising one or more membrane-electrode units as claimed in Claim 29.